



Evaluation of silicon nitride reinforced silica aerogel composites for radom application

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Cite this study: Çetin, İ. E., Başgöz, Ö., Açıkbaz, G., & Açıkbaz, N. Ç. (2022). Evaluation of silicon nitride reinforced silica aerogel composites for radom application. 4th Advanced Engineering Days, 106-110

Keywords

Si₃N₄ aerogel composites
Radom
Aviation
Defense industry
Advanced materials

Abstract

Thanks to its lightness and superior mechanical properties, advanced technology composite materials have a wide range of use in aviation, not only in interiors, but also in the production of structural parts. The radom in the nose of the aircraft is one of the areas of use of advanced technology composite materials. Silica aerogels with high specific surface area (500–1200m²/g), low density (approx. 0.003–0.5 g/cm³), low thermal conductivity (0.005–0.1 Wm⁻¹K⁻¹), ultra-low dielectric constant (1.0–2.0) and low dielectric loss (10⁻² – 10⁻⁴) are potential materials for use in radom application. Recently, with the rapid development of aviation technologies, the speed of aircraft has improved significantly. This situation revealed the necessity of increasing the high temperature performance of silica aerogel radomes. Silicon nitride is a promising structural-functional reinforcement phase for silica aerogel radomes due to its excellent thermo-mechanical and dielectric properties. In this study, studies and results of silica aerogel composites reinforced with Si₃N₄ are presented and discussed.

Introduction

The radomes part of the nose of the aircraft is one of the areas of use of advanced technology materials (Figure 1). The radom is the structure used to protect the antennas from the effects of the outside world. It is an electrically transparent material to electromagnetic energy, where power loss in transmission must be minimal. Therefore, the dielectric constant and loss tangent of the material significantly affect the conduction loss. Radomes have walls separated by a core, as in a solid wall or sandwich structure. The wall configuration of the radom varies according to the application areas. One of the radom types is a graduated porous structure where the porosity lowers the dielectric constant and thus has high power transfer efficiency. This layer can be damaged by moisture due to its porous structure. Radomes are produced from dielectric materials in a way that will least affect the electromagnetic performance of the antennas they contain. In addition, the permeability of radom is often inversely related to its mechanical strength. Carbon has detrimental effects on radom performance because it absorbs electromagnetic radiation and water has a high dielectric constant relative to the radom material itself. The dielectric constant and loss tangent of the material also affect the transmission loss during electromagnetic radiation. In addition to low dielectric constant and loss, high strength, high thermal shock resistance and high thermal stability are the desired basic properties [1].

In radomes, engineering plastics such as glass fiber reinforced epoxy, or ceramic-based composites are used, especially in radomes that will be exposed to high temperatures. The general use temperatures of glass fiber reinforced composites are below 147 °C. In addition, low modulus of elasticity and compressive strength are other factors limiting their use in aviation. The low modulus of elasticity causes excessive stress in the plastic matrix, which causes fractures when stress is applied, thus shortening the fatigue life. Therefore, there is a need for materials that will show high performance, especially at high temperatures [1].



Figure 1. Representation of the radom in the nose of the aircraft [2]

Radomes should be made of materials that can protect internal electronic equipment from high temperatures, are wave-transparent and provide thermal insulation. Silica aerogels with high specific surface area (500–1200m²/g), low density (approx. 0.003–0.5 g/cm³), low thermal conductivity (0.005–0.1 Wm⁻¹ K⁻¹), ultra-low dielectric constant (1.0–2.0) and low dielectric loss (10⁻² – 10⁻⁴) are potential materials for use in this field [3-4-5]. Aerogels are mostly used in the field of thermal insulation due to their low thermal conductivity values, and there are many studies on this subject [6]. However, these applications are limited to non-bearing structures due to fragility [7-8]. Therefore, great efforts are made to produce high-strength aerogel [9]. The incorporation of ceramic fiber into the aerogel not only improves the mechanical properties, but also reduces the radiative heat transfer of the aerogel at high temperature, making it possible to use the aerogel as a load-bearing insulation material as a Thermal Protection System (TPS). It is important to examine the creep behavior of aerogel composite insulation materials with load bearing capacity.

Recently, with the rapid development of aviation technologies, the speed of aircraft has improved significantly. This situation revealed the necessity of increasing the high temperature performance of radomes. Silica aerogels can be used as a radom due to the above-mentioned properties; but their strength is low. In addition, the mesoporous structure of silica aerogels deteriorates above 800°C and the thermal insulation performance deteriorates. It has been reported in the literature that silica aerogels with a density of 0.12 g/cm³ deteriorate under a stress of 31 kPa [10]. In addition, although cross-linked polymer aerogels are suitable for this field, their use is limited in high temperature applications due to thermal pyrolysis of the polymer above 800°C.

To overcome these problems, it is necessary to improve the strength and thermal stability of silica aerogels. There are some methods are reported in literature: Structural strengthening [11-12-13], fiber reinforcement [9, 14-15], polymer crosslinking [16-17-18] or chemical/physical strengthening [19-20-21] procedures have been reported in the literature. Silicon nitride is a promising structural-functional material for radomes due to its excellent thermo-mechanical and dielectric properties [22-23-24-25-26]. By adding Si₃N₄ particles into the aerogel matrix, it is possible to improve the thermo-mechanical properties while maintaining low dielectric constant and low dielectric loss properties.

Si₃N₄ Reinforced Silica Aerogel Composites

In the literature, there are limited studies on the production of Si₃N₄ reinforced silica aerogel composites [27].

Si₃N₄ particle reinforced silica aerogel composites were produced by sol-gel method by drying at ambient pressure by Yang et al. [16]. The microstructure, thermal insulation, mechanical and dielectric properties of the composites were investigated. The effect of the amount of Si₃N₄ (0, 2, 5, 10, 15,20 vol%) on the microstructure and properties is explained. The obtained mesoporous composites were found to have low thermal conductivity (0.024–0.072Wm⁻¹ K⁻¹), low dielectric constant (1.55–1.85) and low dielectric loss (0.005–0.007). As the Si₃N₄ content increased from 5% to 20% by volume, the compressive strength and flexural strength of the composites increased from 3.21 to 12.05MPa and 0.36 to 2.45MPa, respectively. It has been reported that the obtained composites show significant promise in radom applications with the functional integration of wave transparency and thermal insulation. High temperature properties and interface evolution of Si₃N₄ fiber reinforced silica matrix wave transparent composite materials were investigated by another research group [16]. Here the matrix is silica, not aerogel. In the study, Si₃N₄ fiber reinforced silica matrix composites were produced by using the sol-gel method together with the filament winding method in order to improve the high temperature performance of wave transparent materials for high speed aircraft applications. The mechanical properties and interfacial development of the composites at high temperatures were investigated. The properties were evaluated by sintering the composites in two different furnace atmospheres (air and nitrogen). The results showed that composites sintered in a nitrogen atmosphere retained a flexural strength of 210 MPa up to 1200°C, while their air-prepared counterparts held up to approximately 73 MPa. In another study [27], Si₃N₄ particles embedded in the nano-network of silica aerogel prevent aerogel crystallization at high temperatures, thus increasing the strength. In

order to reduce the radiative thermal conductivity, it is additionally doped with the opacifier TiO₂. TiO₂ containing Si₃N₄/SiO₂ aerogel composites were heat treated at 900, 1100, 1200 and 1300 °C for 2 hours. The schematic production process flow chart of the TiO₂ incorporated Si₃N₄/SiO₂ aerogel composites were given in Figure 2.

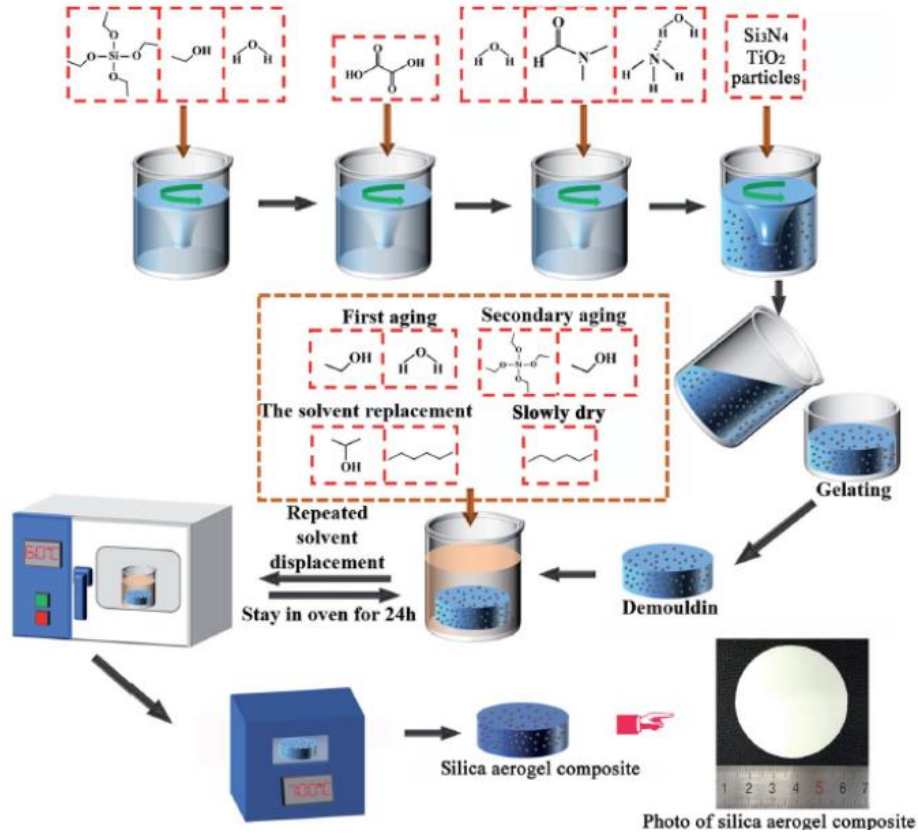


Figure 2. Schematic image of the sample preparation process [27]

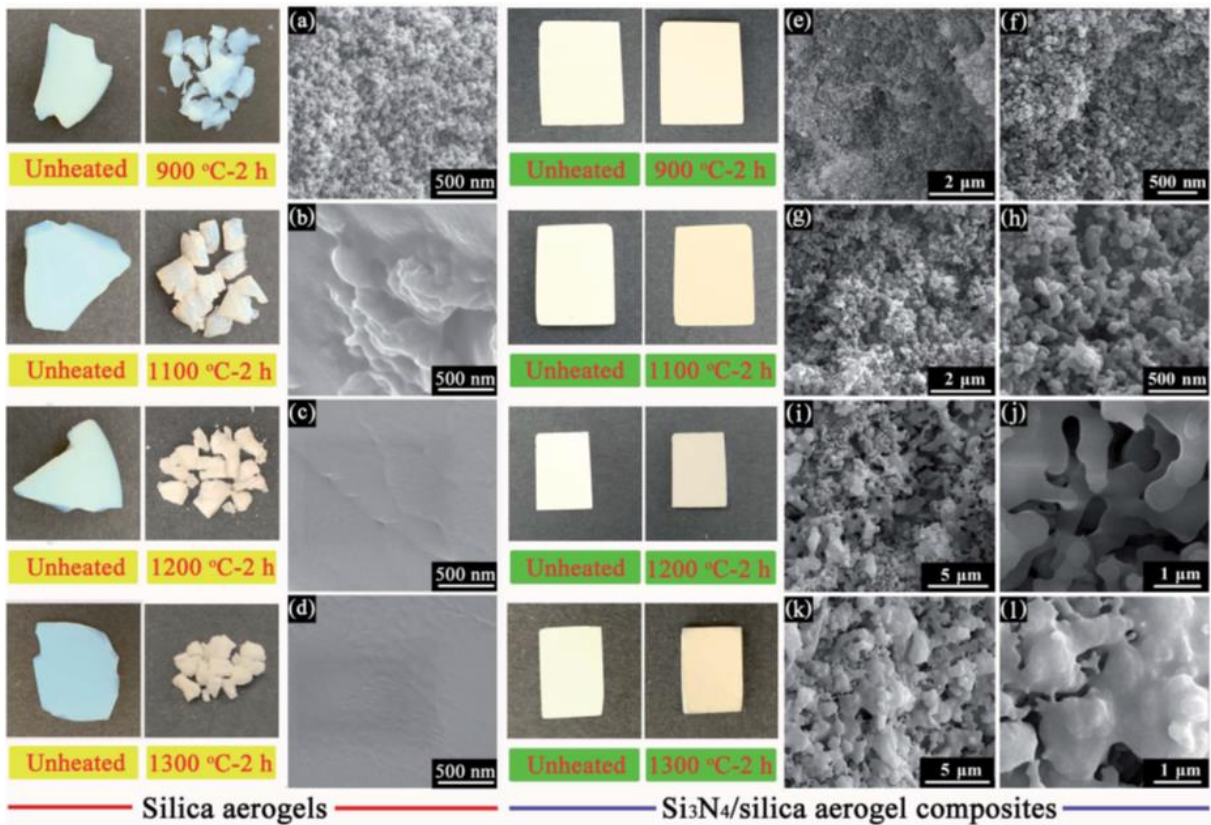


Figure 3. Photographs and SEM images of Si₃N₄-aerogel composite samples heat treated at different temperatures before and after heat treatment [27]

Photographs and SEM images of TiO₂ incorporated Si₃N₄-aerogel composite samples heat treated at different temperatures before and after heat treatment are shown in Figure 3. As seen from the images, after the heat treatment at 900 °C, the pure aerogel sample is brittle. At higher heat treatment temperatures, the silica aerogel within the composite material gradually crystallizes, and the fusion of micro pores causes pore shrinkage and increase in pore size. After heat treatment at 1300 °C, Si₃N₄ particle reinforced composites remained intact without cracks. Si₃N₄ particle addition increased the strength.

Conclusion

When the limited studies in the literature are evaluated, the addition of Si₃N₄ improves the thermo-mechanical properties of silica aerogels. However, there is a need for new production techniques that can be produced on a pilot/large scale so that Si₃N₄ reinforced silica aerogel composites can be used commercially in radom applications.

References

1. Kenion, T., Yang, N., & Xu, C. (2021). Dielectric and Mechanical Properties of Hypersonic Radome Materials and Metamaterial Design: A Review. *Journal of the European Ceramic Society*.
2. <https://ucaklar.org/radom/>
3. Hrubesh, L. W., Keene, L. E., & Latorre, V. R. (1993). Dielectric properties of aerogels. *Journal of materials research*, 8(7), 1736-1741.
4. Jain, A., Rogojevic, S., Ponoth, S., Agarwal, N., Matthew, I., Gill, W. N., ... & Simonyi, E. (2001). Porous silica materials as low-k dielectrics for electronic and optical interconnects. *Thin Solid Films*, 398, 513-522.
5. Maleki, H., Durães, L., & Portugal, A. (2014). An overview on silica aerogels synthesis and different mechanical reinforcing strategies. *Journal of Non-Crystalline Solids*, 385, 55-74.
6. Schmidt, M., & Schwertfeger, F. (1998). Applications for silica aerogel products. *Journal of non-crystalline solids*, 225, 364-368.
7. Parmenter, K. E., & Milstein, F. (1998). Mechanical properties of silica aerogels. *Journal of non-crystalline solids*, 223(3), 179-189.
8. Deng, Z., Wang, J., Wu, A., Shen, J., & Zhou, B. (1998). High strength SiO₂ aerogel insulation. *Journal of non-crystalline solids*, 225, 101-104.
9. Yang, X., Sun, Y., & Shi, D. (2012). Experimental investigation and modeling of the creep behavior of ceramic fiber-reinforced SiO₂ aerogel. *Journal of non-crystalline solids*, 358(3), 519-524.
10. Bertino, M. F., Hund, J. F., Sosa, J., Zhang, G., Sotiriou-Leventis, C., Leventis, N., ... & Terry, J. (2004). High resolution patterning of silica aerogels. *Journal of non-crystalline solids*, 333(1), 108-110.
11. Rao, A.V., S.D. Bhagat, H. Hirashima, G.M. Pajonk, Synthesis of flexible silica aerogels using methyltrimethoxysilane (MTMS) precursor, *J. Colloid Interface Sci.* 300 (2006) 279–285.
12. Nadargi, D. Y., Latthe, S. S., Hirashima, H., & Rao, A. V. (2009). Studies on rheological properties of methyltriethoxysilane (MTES) based flexible superhydrophobic silica aerogels. *Microporous and Mesoporous Materials*, 117(3), 617-626.
13. Aravind, P. R., & Soraru, G. D. (2011). High surface area methyltriethoxysilane-derived aerogels by ambient pressure drying. *Journal of Porous Materials*, 18(2), 159-165.
14. Yuan, B., Ding, S., Wang, D., Wang, G., & Li, H. (2012). Heat insulation properties of silica aerogel/glass fiber composites fabricated by press forming. *Materials Letters*, 75, 204-206.
15. Shi, D., Sun, Y., Feng, J., Yang, X., Han, S., Mi, C., ... & Qi, H. (2013). Experimental investigation on high temperature anisotropic compression properties of ceramic-fiber-reinforced SiO₂ aerogel. *Materials Science and Engineering: A*, 585, 25-31.
16. Yang, H., Kong, X., Zhang, Y., Wu, C., & Cao, E. (2011). Mechanical properties of polymer-modified silica aerogels dried under ambient pressure. *Journal of non-crystalline solids*, 357(19-20), 3447-3453.
17. Guo, H., Meador, M. A. B., McCorkle, L., Quade, D. J., Guo, J., Hamilton, B., ... & Sprowl, G. (2011). Polyimide aerogels cross-linked through amine functionalized polyoligomeric silsesquioxane. *ACS applied materials & interfaces*, 3(2), 546-552.
18. Sabri, F., Marchetta, J., & Smith, K. M. (2013). Thermal conductivity studies of a polyurea cross-linked silica aerogel-RTV 655 compound for cryogenic propellant tank applications in space. *Acta Astronautica*, 91, 173-179.
19. Cai, J., Liu, S., Feng, J., Kimura, S., Wada, M., Kuga, S., & Zhang, L. (2012). Cellulose-silica nanocomposite aerogels by in situ formation of silica in cellulose gel. *Angewandte Chemie*, 124(9), 2118-2121.
20. Boday, D. J., Stover, R. J., Muriithi, B., Keller, M. W., Wertz, J. T., DeFriend Obrey, K. A., & Loy, D. A. (2009). Strong, low-density nanocomposites by chemical vapor deposition and polymerization of cyanoacrylates on aminated silica aerogels. *ACS applied materials & interfaces*, 1(7), 1364-1369.

21. Hu, L., Wang, C. A., & Huang, Y. (2011). Porous YSZ ceramics with unidirectionally aligned pore channel structure: Lowering thermal conductivity by silica aerogels impregnation. *Journal of the European Ceramic Society*, 31(15), 2915-2922.
22. Lukianova, O. A., & Sirota, V. V. (2017). Dielectric properties of silicon nitride ceramics produced by free sintering. *Ceramics International*, 43(11), 8284-8288.
23. Chen, F., Cao, F., Pan, H., Wang, K., Shen, Q., Li, J., & Wang, S. (2012). Mechanical and dielectric properties of silicon nitride ceramics with high and hierarchical porosity. *Materials & Design*, 40, 562-566.
24. Pham, T. A., Li, T., Shankar, S., Gygi, F., & Galli, G. (2011). Microscopic modeling of the dielectric properties of silicon nitride. *Physical Review B*, 84(4), 045308.
25. Acikbas, G., & Calis Acikbas, N. (2021). The effect of sintering regime on superhydrophobicity of silicon nitride modified ceramic surfaces. *Journal of Asian Ceramic Societies*, 9(2), 734-744.
26. Mandal, H., & Acikbas, N. C. (2013). Processing, characterization and mechanical properties of SiAlONs produced from low cost β -Si₃N₄ powder. *KONA Powder and Particle Journal*, 30, 22-30.
27. Yang, H., & Ye, F. (2022). Microtexture, microstructure evolution, and thermal insulation properties of Si₃N₄/silica aerogel composites at high temperatures. *RSC advances*, 12(19), 12226-12234.